

Enhancement of the Upper Critical Field and a Field-Induced Superconductivity in Antiferromagnetic Conductors

Hiroshi Shimahara

Department of Quantum Matter Science, ADSM, Hiroshima University, Higashi-Hiroshima 739-8530, Japan
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We propose a mechanism by which the paramagnetic pair-breaking effect is largely reduced in superconductors with coexisting antiferromagnetic long-range and short-range orders. The mechanism is an extension of the Jaccarino and Peter mechanism to antiferromagnetic conductors, but the resultant phase diagram is quite different. In order to illustrate the mechanism, we examine a model which consists of mobile electrons and antiferromagnetically correlated localized spins with Kondo coupling between them. It is found that for weak Kondo coupling, the superconductivity occurs over an extraordinarily wide region of the magnetic field including zero field. The critical field exceeds the Chandrasekhar and Clogston limit, but there is no lower limit in contrast to the Jaccarino and Peter mechanism. On the other hand, for strong Kondo coupling, both the low-field superconductivity and a field-induced superconductivity occur. Possibilities in hybrid ruthenate cuprate superconductors and some organic superconductors are discussed.

Recently, superconductivity at high fields exceeding the Chandrasekhar and Clogston limit [1,2] (Pauli paramagnetic limit H_P) has been examined by many authors [3–9], in connection with experimental data for organic superconductors [10–15]. In $(\text{TMTSF})_2\text{X}$ and κ -(BEDT-TTF) $_2\text{X}$ compounds in parallel magnetic fields, the upper critical field exhibits an upturn ($d^2H_{c2}/dT^2 > 0$) at low temperatures, exceeding the value of H_P estimated from the zero-field transition temperature $T_c^{(0)}$ by a simplified formula $H_P \sim 1.86 T_c^{(0)}$ [10–14]. On the other hand, in a λ -(BETS) $_2\text{FeCl}_4$ compound, a field-induced superconductivity was observed at high fields, while at zero field it is an antiferromagnetic insulator [15].

There are several possible mechanisms to explain the critical fields higher than H_P . For example, triplet superconductivity, the Fulde-Ferrell-Larkin-Ovchinnikov state (the FFLO state or LOFF state) [16,17], spin-orbit coupling, and a strong coupling effect have been examined. Among them, the upturn of the critical field can be explained by triplet superconductivity with dimensional crossover [7], and also by the FFLO state [3–5]. The pairing symmetries in organic superconductors seem to be still controversial [18].

In triplet superconductors, since the orbital pair-breaking effect is largely suppressed in a parallel magnetic field, the critical field of a parallel spin pairing is much higher than H_P . The upturn and a reentrant superconducting transition are predicted by a theory of dimensional crossover [7].

The microscopic origin of the triplet superconductivity has been studied by many authors. For example, on the analogy of the superfluid ^3He , a paramagnon theory based on the ferromagnetic fluctuation appears to be a natural explanation, but many of the organics are in proximity to the antiferromagnetic phase. This discrepancy does not exist in phonon mechanisms of the triplet superconductivity. The anisotropic components of the pairing interactions are rather large due to the weak

screening effect in the layered systems [19]. Hence, reduction of the s -wave pairing interaction due to short-range Coulomb repulsion may give rise to triplet superconductivity [19,20].

The pairing interactions mediated by antiferromagnetic fluctuations may also be a mechanism of triplet superconductivity. The pairing interactions mediated by antiferromagnetic fluctuations include attractive triplet channels [21]. Hence, suppression of the singlet superconductivity due to some additional mechanism, such as inter-site Coulomb repulsion or a magnetic field (if it is applied), may give rise to triplet superconductivity [21].

On the other hand, in singlet superconductors, the FFLO state is a candidate for the mechanism of the high-field superconductivity and the upturn of the upper critical field. Recently, it was observed in κ -(BEDT-TTF) $_2\text{Cu}[\text{N}(\text{CN})_2]\text{Br}$ that the upturn has a tendency to disappear when the direction of the magnetic field is slightly tilted [22]. This behavior is consistent with a theoretical prediction based on the FFLO state. To obtain direct evidence of the FFLO state, observation of the spatial structure of the gap function by scanning tunneling spectroscopy (STS) would be useful [4,5]. Purely quantitative arguments for the critical field are difficult because detailed information on the Fermi surface structure and the interactions is necessary for accurate estimation [23,9,24]. Subdominant interactions presumably exist in anisotropic superconductors [19,21].

Furthermore, it should be noted that in strong coupling superconductors, the observed critical fields are larger than the values of H_P expected from $T_c^{(0)}$ in weak coupling theory, because the ratio $T_c^{(0)}/\Delta_0$ is reduced by a strong coupling effect, where Δ_0 is the BCS gap at $T = 0$ and $H = 0$.

Jaccarino and Peter proposed a mechanism of ultrahigh-field superconductivity [25]. They pointed out that the exchange field created by the polarized rare earth spin cancels the magnetic field in cer-

tain ferromagnetic metals. The Jaccarino and Peter mechanism explains the field-induced superconductivity in $\text{Eu}_x\text{Sn}_{1-x}\text{Mo}_6\text{S}_8$ [26] and probably that in the λ -(BETS) $_2\text{FeCl}_4$ compound [15]. An observed temperature dependence of the lower critical field of the high-field superconductivity in λ -(BETS) $_2\text{FeCl}_4$ [27] can be explained by the FFLO state in combination with the Jaccarino and Peter mechanism.

In this paper, we propose a mechanism by which the paramagnetic pair-breaking effect is largely reduced in superconductors with coexisting antiferromagnetic long-range and short-range orders. The present mechanism is an extension of the Jaccarino and Peter mechanism of ferromagnetic metals to antiferromagnetic metals. The origin of the enhancement of the critical field is due to a compensation effect similar to that of the Jaccarino and Peter mechanism. However, the resultant phase diagram is quite different for low fields or weak exchange fields. Possible candidates of the present mechanism may be found in some of the layered superconductors in proximity to the antiferromagnetic phase.

In order to describe the mechanism, we divide the system into a conductive electron system and a magnetic subsystem of localized spins. We introduce an antiferromagnetic exchange coupling between the localized spins, and Kondo coupling between the electrons and the spins. This model is called a generalized Kondo lattice model and a spin fermion model. We consider a multilayer structure and parallel magnetic fields, in which the orbital pair-breaking effect is suppressed.

When the magnetic field is applied to the antiferromagnetic long-range order, a canted spin structure occurs. Then, the ferromagnetic moment is induced in the spin subsystem, and it creates the exchange field on the mobile electrons, which cancels the magnetic field in the Zeeman energy term.

In addition to the exchange fields, the localized spins create the internal magnetic fields, which cause the Lorentz force in the mobile electrons. We ignore this additional internal field for simplicity and to concentrate on the spin effect. This simplification is quantitatively correct in the case where magnetic layers and conductive layers are spatially separated. For example, in $\text{RuSr}_2\text{GdCu}_2\text{O}_8$ compounds, the ruthenate layers are distant from the cuprate layers, and the internal magnetic field is considered to be weak [28].

We assume the long-range order for simplicity, but the same effect is expected for the short-range order. Within the antiferromagnetic correlation length, the cancellation effect occurs. The fluctuations of the correlated spins do not change the direction of the exchange field, which is always opposite to the magnetic field.

First, we consider canted spin structures of the localized spin system. In reality, the localized spin state is modified by the mobile electron state, and the total state should be determined self-consistently [29]. However, we assume that such modifications are already included in the spin Hamiltonian considered below, and

that the canted spin structure exists as a renormalized state. These assumptions are qualitatively appropriate for the systems with strong antiferromagnetic correlations.

The spin Hamiltonian is defined by

$$\mathcal{H}_J = J \sum_{(i,j)} \mathbf{S}_i \cdot \mathbf{S}_j - 2\mu_0 \sum_i \mathbf{S}_i \cdot \mathbf{H}, \quad (1)$$

where \mathbf{H} is the magnetic field. Here, μ_0 is the magnetic moment of the electron, such that $2\mu_0 = -g\mu_B < 0$ with the Bohr magneton μ_B and the g -factor. We define sublattices A and B . The spin coordinate is defined in two ways for convenience. We take the z -axis in the direction perpendicular to the layers, and the x and y -axes parallel to the layers. For convenience in consideration of the localized spin states, we take z', x', y' -axes along the x, z, y -axes, respectively. We consider the magnetic field applied in the $-x$ direction parallel to the layers, that is $\mathbf{H} = (H, 0, 0)$ with $H < 0$.

We assume the classical spins of the magnitude S for simplicity. This treatment is equivalent to the mean field theory at low temperatures, when S is replaced by the spin moment. We define the polar coordinate (θ'_i, φ'_i) to express the direction of the classical spin on site i in the spin coordinate system $x'y'z'$, that is, $\langle \mathbf{S}_i \rangle = S (\sin \theta'_i \cos \varphi'_i, \sin \theta'_i \sin \varphi'_i, \cos \theta'_i)$. When we consider quantum spins, the influence of the shrinkage of the spins due to the fluctuations can be partially taken into account by the replacement of S with $M = |\langle \mathbf{S}_i \rangle|$. The energy of the localized spin part $\langle \mathcal{H}_J \rangle \equiv E_J$ is written as

$$E_J = JS^2 \sum_{(i,j)} (\sin \theta_i \sin \theta'_j \cos(\varphi'_i - \varphi'_j) + \cos \theta'_i \cos \theta'_j) - 2\mu_0 HS \sum_i \cos \theta'_i, \quad (2)$$

which becomes minimum when $\varphi'_i = \varphi'_j + \pi$ for $i \in A$ and $j \in B$, and $\theta'_i = \theta$ for any i , where θ is an angle such that

$$\cos \theta \equiv \frac{\mu_0 H}{zJS} \quad (3)$$

for $|\mu_0 H| \leq zJS$, and $\theta = 0$ for $|\mu_0 H| > zJS$. The minimum value is $E_J = -zN_s JS^2 [1 + 2(\mu_0 H/zJS)^2]/2$ for $|\mu_0 H| \leq zJS$, and $E_J = -zN_s JS^2 [-1 + 4\mu_0 H/zJS]/2$ for $|\mu_0 H| > zJS$. Here, z and N_s are the number of nearest neighbor sites and the total number of lattice sites, respectively.

Next, we consider the effective Hamiltonian for the mobile electrons, which is defined by

$$\mathcal{H}_e = \mathcal{H}_0 + \mathcal{H}_K \quad (4)$$

with

$$\begin{aligned} \mathcal{H}_0 = & -t \sum_{(i,j),\sigma} c_{i\sigma}^\dagger c_{j\sigma} - \mu \sum_{i,\sigma} c_{i\sigma}^\dagger c_{i\sigma} \\ & - \sum_{i,\sigma_1,\sigma_2} \mu_0 \mathbf{H} \cdot (c_{i\sigma_1}^\dagger \vec{\sigma}_{\sigma_1\sigma_2} c_{i\sigma_2}) \end{aligned} \quad (5)$$

and

$$\mathcal{H}_K = J_K \sum_{i, \sigma_1, \sigma_2} \mathbf{S}_i \cdot (c_{i\sigma_1}^\dagger \vec{\sigma}_{\sigma_1 \sigma_2} c_{i\sigma_2}). \quad (6)$$

Here, we have omitted pairing interaction terms in eq. (4) [30], since our purpose is to examine the modification of the electron dispersion relation by the exchange fields. In terms of the interlayer hopping energy t_\perp and the on-site Coulomb repulsion on the magnetic layers U_m , the Kondo coupling is written as $J_K = t_\perp^2/U_m > 0$ within a second-order perturbation theory.

In the background of the canted spin structure of the magnetic layers, \mathcal{H}_K is rewritten as

$$\begin{aligned} \mathcal{H}_K = & J_K S \cos \theta \left[\sum_{i \in A} (a_{i\uparrow}^\dagger a_{i\downarrow} + a_{i\downarrow}^\dagger a_{i\uparrow}) \right. \\ & \left. + \sum_{j \in B} (b_{j\uparrow}^\dagger b_{j\downarrow} + b_{j\downarrow}^\dagger b_{j\uparrow}) \right] + J_K S \sin \theta \\ & \times \left[\sum_{i \in A, \sigma} \sigma a_{i\sigma}^\dagger a_{i\sigma} - \sum_{j \in B, \sigma} \sigma b_{j\sigma}^\dagger b_{j\sigma} \right], \end{aligned} \quad (7)$$

where we have defined the electron operators $a_{i\sigma}$ and $b_{j\sigma}$ on the sites $i \in A$ and $j \in B$. Here, we find that the weak ferromagnetic moment in the x direction ($S \cos \theta > 0$) increases the population of spins in the $-x$ direction when $J_K > 0$. Furthermore, \mathcal{H}_0 is also rewritten as

$$\begin{aligned} \mathcal{H}_0 = & -t \sum_{\substack{(i,j), \sigma \\ i \in A, j \in B}} (a_{i\sigma}^\dagger b_{j\sigma} + b_{j\sigma}^\dagger a_{i\sigma}) - \mu \left[\sum_{i \in A, \sigma} a_{i\sigma}^\dagger a_{i\sigma} \right. \\ & \left. + \sum_{j \in B, \sigma} b_{j\sigma}^\dagger b_{j\sigma} \right] - \mu_0 H \left[\sum_{i \in A} (a_{i\uparrow}^\dagger a_{i\downarrow} + a_{i\downarrow}^\dagger a_{i\uparrow}) \right. \\ & \left. + \sum_{j \in B} (b_{j\uparrow}^\dagger b_{j\downarrow} + b_{j\downarrow}^\dagger b_{j\uparrow}) \right]. \end{aligned} \quad (8)$$

Therefore, we obtain an expression

$$\mathcal{H}_e = \sum_{\mathbf{k}}' \left(a_{\mathbf{k}\uparrow}^\dagger b_{\mathbf{k}\uparrow}^\dagger a_{\mathbf{k}\downarrow}^\dagger b_{\mathbf{k}\downarrow}^\dagger \right) \mathcal{E}_{\mathbf{k}} \begin{pmatrix} a_{\mathbf{k}\uparrow} \\ b_{\mathbf{k}\uparrow} \\ a_{\mathbf{k}\downarrow} \\ b_{\mathbf{k}\downarrow} \end{pmatrix} \quad (9)$$

with 4×4 matrix $\mathcal{E}_{\mathbf{k}}$ defined by

$$\mathcal{E}_{\mathbf{k}} = \begin{pmatrix} -\mu - h_z & \epsilon_{\mathbf{k}} & -h_x & 0 \\ \epsilon_{\mathbf{k}} & -\mu + h_z & 0 & -h_x \\ -h_x & 0 & -\mu + h_z & \epsilon_{\mathbf{k}} \\ 0 & -h_x & \epsilon_{\mathbf{k}} & -\mu - h_z \end{pmatrix} \quad (10)$$

and

$$\begin{aligned} \epsilon_{\mathbf{k}} & \equiv -2t (\cos k_x + \cos k_y) \\ h_x & \equiv \mu_0 H - J_K S \cos \theta \\ h_z & \equiv -J_K S \sin \theta, \end{aligned} \quad (11)$$

where \mathbf{k} and $\sum_{\mathbf{k}}'$ are the sublattice momentum and the summation over the first Brillouin zone of the sublattice

momentum space, respectively. The lattice constants are taken as unity.

The eigenvalues of the matrix $\mathcal{E}_{\mathbf{k}}$ are expressed as

$$\tilde{\epsilon}_{\mathbf{k}} = -\mu \pm \left[(\epsilon_{\mathbf{k}} \pm h_x)^2 + h_z^2 \right]^{1/2}. \quad (12)$$

If the system is away from the half-filling and has any pairing interactions, superconductivity occurs in the renormalized electron system with the energy dispersion relation described by eq. (12) [31].

The first plus or minus sign \pm in eq. (12) corresponds to the upper and lower bands which are divided by the exchange field in the z direction. The band gap does not affect the superconducting transition when the system is away from the half-filling, whereas near the half-filling, the mobile electron layer becomes an insulator and the superconductivity does not occur when $\theta \neq 0$, i.e., $|\mu_0 H| < zJS$.

The second plus or minus sign \pm corresponds to the split of the Fermi surfaces of up and down spin electrons when the x -axis is taken as the quantization axis of the spin space. This split causes the pair-breaking effect, and as a consequence the upper critical field is bounded by the Pauli paramagnetic limit. However, as shown in eq. (11), the magnitude of h_x can be smaller than $|\mu_0 H|$ due to the cancellation of $\mu_0 H$ and the exchange field $J_K S \cos \theta$. From eqs. (3) and (11), the effective field H_{eff} , such that $h_x = \mu_0 H_{\text{eff}}$, is

$$H_{\text{eff}} = H \left(1 - \frac{J_K}{zJ} \right) \quad (13)$$

for $|\mu_0 H| \leq zJS$, and $H_{\text{eff}} = H - \text{sign}(H) J_K S / |\mu_0|$ for $|\mu_0 H| > zJS$.

The superconductivity occurs when $|H_{\text{eff}}| < H_c^{(0)}(T)$, where $H_c^{(0)}(T)$ is the upper critical field (H_P or an FFLO critical field) in the absence of the exchange field at a temperature T . Here, we assume that the antiferromagnetic transition occurs at a higher temperature, and the magnetic order can be regarded as a rigid background. Therefore, for $|\mu_0 H| \leq zJS$, the superconductivity occurs when

$$|H| < H_c \equiv \frac{H_c^{(0)}}{|1 - J_K/zJ|}. \quad (14)$$

Thus, H_c is the critical field when $J_K S < zJS - \mu_0 H_c^{(0)}$ or $J_K S > zJS + \mu_0 H_c^{(0)}$. On the other hand, for $|\mu_0 H| \geq zJS$, the superconductivity occurs when

$$\frac{J_K S}{|\mu_0|} - H_c^{(0)} < |H| < \frac{J_K S}{|\mu_0|} + H_c^{(0)}. \quad (15)$$

Therefore, we obtain the phase diagram shown in Fig. 1. We have chosen the parameter $|\mu_0| H_c^{(0)} / zJS = 0.2$ as an example, which is consistent with our assumption that the magnetic long-range or short-range order exists at the superconducting transition temperature. It

is found that when $J_K S < zJS - |\mu_0|H_c^{(0)}$ the upper critical field is enhanced by the factor $1/(1 - J_K/zJ) > 1$, but a field-induced transition is not obtained, in contrast to the Jaccarino and Peter mechanism. When $J_K \sim zJ$, the critical field reaches a value of the order of $H_c \sim zJS/|\mu_0| \sim T_{AF}^*/|\mu_0|$. Here, T_{AF}^* is a crossover temperature at which antiferromagnetic fluctuations begin to occur. In the absence of low-dimensional thermal fluctuations, T_{AF}^* is of the order of the antiferromagnetic transition temperature.

For strong Kondo coupling $J_K S > zJS + |\mu_0|H_c^{(0)}$, we find a reentrant transition to a high-field phase in addition to a low-field phase. For the high fields $|\mu_0 H| > zJS$, the present mechanism coincides with the original Jaccarino and Peter mechanism, since the spin moments are saturated.

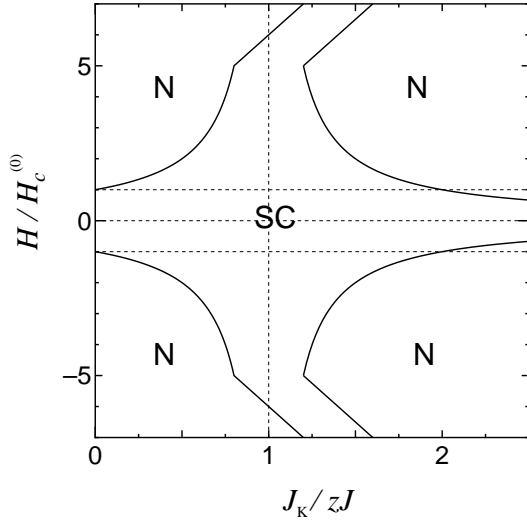


FIG. 1. Phase diagram on the J_K/zJ and $H/H_c^{(0)}$ plane when $|\mu_0|H_c^{(0)}/zJS = 0.2$. The signs N and SC denote the normal state and the superconductivity, respectively.

In conclusion, we find that the upper critical field is enhanced by the coupled antiferromagnetic layers due to a cancellation effect of the external field and the exchange field induced by the canted spin structure. In particular, when $J_K \sim zJ$, the system is almost free from the paramagnetic pair-breaking effect for practical strengths of the magnetic field. The critical field reaches a value of the order of $T_{AF}^*/|\mu_0|$. If such an antiferromagnetic quasi-two-dimensional metal is synthesized and exhibits superconductivity, it can be a superconductor with an extraordinarily high critical field.

The present phase diagram is very different from that of the original Jaccarino and Peter mechanism for ferromagnetic metals. It is found that for small J_K , a field-induced superconductivity does not occur, whereas for large J_K , both the field-induced superconductivity and the low-field superconductivity occur.

In the phase diagram, the metal-insulator transition is not presented. However, for example, at the half-filling, the system is an insulator with a canted spin structure when $|\mu_0 H| < zJS$. In this case, the phase diagram for large J_K is similar to that in the λ -(BETS) $_2$ FeCl $_4$ compound [15,32]. The spin moments on FeCl $_4$ would create an exchange field on the conduction hole band of the two-dimensional network of the BETS molecules. The hybrid ruthenate cuprate compounds are also possible candidates due to their crystal structures.

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 - [32] *Note added in proof:* In this paper, we have additionally discussed a possibility of the FFLO state in combination with the Jaccarino and Peter mechanism in λ -(BETS)₂FeCl₄. Recently, Tanatar *et al.* reported thermal conductivity data that support the FFLO state in λ -(BETS)₂GaCl₄ [M. A. Tanatar, T. Ishiguro, H. Tanaka and H. Kobayashi: preprint].